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COMPLETE SPECIFICATION

Improvements in or relating to Expandable Polystyrene

We, KOPPERS COMPANY, INC., a corporation organized under the laws of the State of Delaware, one of the United States of America, of 436 Seventh Avenue, City of Pittsburgh, Commonwealth of Pennsylvania, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates generally to expandable cellular polymeric materials and more particularly to expandable polystyrene compositions suitable for producing foams having improved surface appearance, solvent resistance, and water vapor transmission resistance.

The advent of expandable polymeric materials, and particularly expandable polystyrene, has been a valuable addition to the art of container and packaging manufacture because expanded polystyrene foams are greatly superior in impact resistance and thermal insulation qualities to other container and packaging materials. Consequently, expanded foams prepared from expandable polystyrene offer unique advantages in the manufacture of lightweight, thermally insulated beverage containers and impact resistant packages.

Expandable polystyrene is commercially available as polymeric particles in bead or pellet form which contain a blowing agent, such as a hydrocarbon, dispersed within the particles. These particles which contain hydrocarbon blowing, or expanding, agents can be processed by extrusion, blow molding or injection molding to produce a shaped foam container or package. Under the influence of heat using any of the above-described techniques, the blowing agent causes the polymer particles to expand and fuse together forming the final foam product. The final product, characteristically, is in the nature of small discrete cells fused or bonded together.

To date, these products manufactured from expandable polystyrene exhibit a surface skin which tends to show the line of junction of the discrete particles and most liquids used in the containers will stain the surface of the foamed product so as to outline these boundary lines or cells. While the skin of the expandable polystyrene products is generally tough it is extremely susceptible to abrasion and the surface itself has a dull non-aesthetic appearance. Another disadvantage of containers or products made from expanded polystyrene foams is that they have unsatisfactory water vapor transmission characteristics. The problem created by this disadvantage is serious inasmuch as the thermally insulating expanded polystyrene containers are designed primarily for use as, for example, ice buckets, cups or trays to contain heated or cooled beverages.

Staining of the foam containers by whatever beverage may be contained therein does not permit more than a single use for each container. Additionally, the containers cannot retain beverages for a great length of time due to the rapid water vapor transmission characteristics. The containers are also limited to the type of liquid which may be used therein. For example, materials which indicate even a slight solvent action on polystyrene cannot be packaged and shipped in containers made of expandable polystyrene because the polystyrene material will not resist sufficiently this solvent action and the liquids would have a tendency to leak from these containers.

Expandable polystyrene has found wide use in packaging of fragile instruments, such as typewriters and glass. However, there are also many delicate instruments which are shipped in containers and must be kept relatively free from moisture. Because of the water vapor transmission characteristics of expanded polystyrene foam they cannot advantageously be used to ship instruments which must be kept within relatively dry atmospheres. Consequently, the full value of the expanded polystyrene foams cannot be realized.

We have found that if powdered polyethylene is incorporated into expandable polystyrene compositions the foam materials pro-

duced therefrom possess an improved surface appearance, an improved solvent resistance and a heretofore unknown resistance to the transmission of water vapor.

According to the invention there is provided an expandable thermoplastic composition capable of being molded into a foam structure which comprises a substantially uniform mixture of (a) particles of polystyrene having homogeneously incorporated therein a blowing agent boiling below the incipient softening point of the polystyrene particles and (b) polyethylene, the quantity of polystyrene in the composition exceeding the quantity of polyethylene in the composition.

Advantageously, the composition takes the form of extruded pellets and the polyethylene is incorporated within the extruded pellets.

The expandable polystyrene particles may be prepared by any of the conventional methods known in the art. For example, expandable polystyrene beads have been produced by the suspension polymerization method; in this method suspended beads of polystyrene are subjected to an atmosphere of a selected blowing agent whereby the beads are impregnated with the blowing agent. Alternatively, the suspension polymerization may be conducted in the presence of the blowing agent so that the polymer beads contain upon conclusion of the polymerization reaction a stated percentage of the blowing agent. In many cases, it will be advantageous to incorporate in the composition a normally solid two component blowing agent adjunct of which one component is boric acid or a normally solid organic acid containing at least three milliequivalents of acidic hydrogen per gram and the other component is a carbon dioxide-liberating agent, the two components being capable of reacting together to form carbon dioxide and water. The practice of adding such a two-component blowing agent adjunct is described in detail in Specifications Nos. 783,034 and 994,073.

A preferred method of preparing the expandable polystyrene particles is described in Specification No. 994,074. This method is known as the quench pellet method; in this method, a normally liquid blowing agent is homogeneously and uniformly admixed with the polystyrene resin that is in a plastic state under conditions such that foaming of the resin does not occur, the admixture extruded through a die of the desired shape and the admixture then quenched before foaming can occur. The admixture, after quenching, is pelletized to render it suitable for fabrication in a molding process or for extruding processes such as are useful in the method of this invention.

Compositions in accordance with the present invention may be formed into a foam structure having a smooth continuous surface appearance, a resistance to the action of solvents and a resistance to permeation of the foam structure by moisture. Accordingly, these compositions can be charged into a closed mold, and subjected to heat of sufficient temperature to soften the composition material and cause expansion of the blowing agent to expand the mass of pellets into an integral molded foam structure. Additionally, the composition in the form of pellets may be blow-molded into shapes, such as bottles, using conventional blow-molding equipment.

The results achieved by this invention are surprising in view of the fact that polyethylene and polystyrene are incompatible materials, that is the materials are known to be, to a great extent, insoluble in each other. Thus, polyethylene and polystyrene not being mutually soluble, a composition exhibiting properties such as those of this invention is totally unexpected.

Articles produced from the compositions of this invention are comprised of foam structures of expanded polystyrene material containing a high-density polyethylene homogeneously dispersed throughout, which structures have a smooth uniform surface and which structures possess water and solvent-resistant characteristics which have been heretofore unknown and unobtainable when produced from regular expandable polystyrene particles or pellets.

Polyethylene materials of varying powder sizes may be employed in the practice of this invention. For example, a mixture of high density polyethylene powder of the following screen analysis may be advantageously used to produce the novel composition of this invention.

SCREEN ANALYSIS OF POLYETHYLENE POWDER

High Density Polyethylene Powder
Melt Index = 13.6 gm/10 min.

Through-On	Per Cent
— 40 mesh[1]	2.4
40 mesh — 80 mesh	2.8
80 mesh — 100 mesh	2.2
100 mesh — 140 mesh	7.6
140 mesh — 200 mesh	11.5
200 mesh — 325 mesh	56.2
325 mesh	17.3

[1] U.S. Standard Mesh

It should be understood that this particular combination of polyethylene powder of this melt index is not the only combination operable in this invention.

Preferably, the polyethylene powder material is incorporated within the expandable polystyrene particles during the formation or preparation of the expandable particles. By way of example, the polystyrene, polyethylene powder and blowing agent can be combined in the feed hopper of an extruder in sufficient quantity so that at the desired feed rate the feed worms will be full. The polystyrene may

contain some of the blowing agent, all of the blowing agent, or none of the blowing agent to be contained in the final composition. A pressure generated by the feed worms forces the particles into the extruder mixing zone where, in those instances where the polystyrene contains none or only a small proportion of the blowing agent, the or additional blowing agent may be forced into the plasticized mass by means of a constant delivery pressure pump. The action of the screw forcing the plastic mass through the barrel of the extruder causes the blowing agent to become intimately and homogeneously admixed with the plastic mass. The blowing agent may also be injected into the plastic mass through the screw rather than, or in addition to, injection through the barrel.

The extruder used in the preparation of the compositions of this invention is generally of conventional design. Suitable extruders are described in chapter 4 of "Processing of Thermoplastic Materials" edited by E. C. Barnhardt, 1959. Depending upon screw design, provision may be made to prevent feedback of the blowing agent into the feed zone of the hopper. For example, if the blowing agent is a hydrocarbon, it may tend to flow back toward the hopper and condense out on the beads causing them to cake and thus prevent continuous operation. This tendency can be avoided by proper screw design and addition of the blowing agent to the extruded mass at the proper place. The extruder is operated at temperatures such that the rendering of the polymer to the plastic state takes place rapidly thereby inhibiting the escape of the blowing agent. Typical extrusion temperatures are:

Rear cylinder temperatures	240—250°F.
Front cylinder temperatures	255—270°F.
Die temperatures	270—290°F.

The plastic mass containing the blowing agent is forced through the die of desired shape such as a strand die.

The polymer mass leaving the extruder die is quenched before foaming can occur. By quenching is meant cooling at a rapid rate, e.g. as quickly as possible, from the extrusion temperature to a temperature lower than the softening temperature of the polymer. The quenching of the extrudate should immediately follow the extrusion, preferably without permitting the extrudate to be exposed to the atmosphere for any substantial length of time. This can usually be accomplished by extruding directly into a cooling bath, e.g. water, or by using a specially constructed die wherein the exit end of the die is insulated from the heated portion of the die and is cooled, such as by circulating a liquid or gas refrigerant through said die.

The quantity of polyethylene material which may be employed in the total composition depends upon a variety of factors and conditions. For example, the quantity of polymeric particles being expanded to produce a foamed object and the desired characteristics of the end products formed will dictate the ultimate quantities. Ordinarily, it is preferred to use enough of the polyethylene powder to ensure improvements in vapor transmission characteristics, solvent resistance and surface appearance. In this respect at least one per cent by weight of the powdered polyethylene is desirable in the composition of this invention. However, in any case the polystyrene contained in the composition should always exceed, in quantity, the powdered polyethylene.

The use of normally liquid blowing agents in amounts of up to 15 parts per 100 parts of polymeric particles, that is polystyrene, plus polyethylene, plus expanding agent, yields usable expandable polymeric particles. Particularly good results are obtained by the use of about seven parts of blowing agent per 100 parts of polymer particles, but greater or lesser amounts may be used depending upon the use to which the resulting particles will be put.

Among the blowing agents useful in this invention are the normally liquid aliphatic hydrocarbons boiling in the range of 20—100°C. and mixtures thereof with volatile solvents in such amounts that the resulting mixture is not a solvent for the polymer. More generallly, aliphatic hydrocarbons boiling below the incipient softening point of the polymer with which they are to be used are useful in the process of this invention. Excellent results are obtained with blowing agents boiling in the range 25—65°C.

The method of the present invention is further illustrated by the following Example.

EXAMPLE

Expandable polystyrene beads (sold under the trade-mark Dylite), containing 7.75 per cent by weight of normal pentane, in an amount of 95 parts by weight was charged to the hopper of a National Rubber Machine Company 1.5 inch extruder having a length to diameter ratio of 20:1 while simultaneously five parts by weight of a powdered polyethylene of the following screen analysis was charged.

Through-On	Per Cent
— 40 mesh[1]	2.4
40 mesh — 80 mesh	2.8
80 mesh — 100 mesh	2.2
100 mesh — 140 mesh	7.6
140 mesh — 200 mesh	11.5
200 mesh — 325 mesh	56.2
325 mesh	17.3

[1] U.S. Standard Mesh

The extruder was started and run using the following conditions:

Screw speed	50 r.p.m.
Rear cylinder temperature	245°F.
Front cylinder temperature	265°F.
Die temperature	280°F.

The die which contains nine, one-sixteenth inch diameter ports was immersed in a water bath which was held at 54°F. The extruded strands travelled for 20 inches under the water at a rate of 25 linear feet per minute. The strands were withdrawn from the water bath and passed into a strand pelletizer by powered rollers. The strands of the extrudate were chopped into pellets of about three-sixteenth inch length and thereafter stored in containers at ambient room conditions.

A portion of these pellets were then mixed with a multi-component blowing agent adjunct comprised of 0.30 per cent citric acid monohydrate and 0.22 per cent sodium bicarbonate until said pellets were externally coated with said adjunct. The adjunct is particularly advantageous for use with pellets that are to be processed in an extruding operation to obtain lower density foams, e.g. 2.5—5 pounds per cubic foot. These pellets containing the externally added adjunct were extrusion blow molded in conventional extrusion blow molding equipment to produce expanded polystyrene-polyethylene composition materials in the shape of gallon bottles.

A separate portion of the quenched pellets produced in the Example was placed into a closed mold and subjected to the action of steam and molded into the form of a board.

Visual inspection of the extruded and steam molded materials indicated that they had a smooth uniform surface noticeably absent of any visual line of junction between the discrete particles. Additionally, moisture vapor permeability data and solvent resistant data were gathered on the products thus produced. The foregoing procedure was repeated for several runs except that the amount of powdered polyethylene used in the composition was varied. Results of the tests performed on all the materials are reproduced in Table I.

By way of comparison when the foregoing procedure was repeated exactly, with the exception that the polyethylene powder was not incorporated in the materials and extrusion blow molded and steam molded structures were prepared, clearly defined juncture lines appeared on the surfaces. In addition the solvent resistance and moisture vapor transmission resistance were inferior to the extrusion and steam molded foam structures obtained with the use of the combined polystyrene-polyethylene material. These results are also reproduced in Table I as run numbers 2 and 6.

TABLE I

Physical Property Evaluation of Blow Molded Bottles and Steam Molded Blocks

Run No.	Material	Composition of Quenched Pellet		Extrusion Stock Temperature °F.	Steam Molding Pressure (Pounds)	Water Vapor Transmission in Perms (3) on Thickness as Shown (Transmission Estimated From 72-Hour Evaluation)		Solvent Resistance Estimate Disintegration Time in Benzene (2)
		Expandable Polystyrene	Polyethylene Powder %			Perms	Thickness	
1	Quenched Pellet (1)	99	1	220		1.4	115 mils	2 minutes
2	Quenched Pellet (1)	100	0	220		2.0	115 mils	4 seconds
3	Quenched Pellet (1)	97	3	220		0.8	110 mils	4 minutes
4	Quenched Pellet (1)	95	5	220		0.4	115 mils	55 minutes
5	Quenched Pellet (1)	90	10	220		0.4	114 mils	Greater than 4 hours
6	Quenched Pellet	100			12—15	2.1	1 inch	4 minutes (0.7 ml)
7	Quenched Pellet	90	10		12—15	0.38	1 inch	41 minutes (12.8 ml)
8	Quenched Pellet	95	5		12—15	0.4	1 inch	39 minutes
9	Quenched Pellet	97	3		12—15	0.7	1 inch	29 minutes

(1) 0.30 per cent citric acid monohydrate and
0.02 per cent sodium bicarbonate were added externally to pellets before they were extrusion blow molded.

(2) Benzene was added dropwise at a constant rate onto the specimen until the liquid was observed on the underside of this specimen. The permeation time, an arbitrary measure of solvent resistance, represents the elapsed time from the first addition (drop) until benzene was seen on the underside of the testing area.

(3) A.S.T.M. Test Method E96—63T.

As is readily apparent the present invention permits superior foam molded articles to be manufactured having greater efficiencies with respect to surface appearance, vapor transmission and solvent resistance. These benefits are obtained from the improved composition of expandable material prepared in accordance with this invention.

WHAT WE CLAIM IS:—

1. An expandable thermoplastic composition capable of being molded into a foam structure, which comprises a substantially uniform mixture of (a) particles of polystyrene having homogeneously incorporated therein a blowing agent boiling below the incipient softening point of the polystyrene particles and (b) polyethylene, the quantity of polystyrene in the composition exceeding the quantity of polyethylene in the composition.

2. A composition as claimed in Claim 1, wherein the thermoplastic composition is in the form of extruded pellets.

3. A composition as claimed in Claim 2, wherein the polyethylene is incorporated within the extruded pellets.

4. A composition as claimed in any one of the preceding claims, wherein said polyethylene constitutes at least one per cent by weight of the total composition.

5. A composition as claimed in any one of the preceding claims, wherein said blowing agent is an aliphatic hydrocarbon boiling in the range of from 20°C. to 100°C.

6. A composition as claimed in Claim 1, substantially as described in the foregoing Example.

7. A method of preparing an expandable thermoplastic composition which comprises passing polystyrene, powdered polyethylene, the quantity of polystyrene used exceeding the quantity of polyethylene used, and a blowing agent boiling below the incipient softening point of the polystyrene through a screw extruder in which said thermoplastic polystyrene and polyethylene become molten, extruding the mixture of said molten thermoplastic polystyrene and polyethylene and blowing agent at a temperature greater than the softening temperature of said thermoplastics before said thermoplastics have had an opportunity to expand, and quenching the extruded mixture to a temperature less than the softening temperature of said thermoplastics.

8. A method according to Claim 7, wherein said polystyrene contains said blowing agent prior to introduction into the extruder.

9. A method of preparing an expandable thermoplastic composition according to Claim 7, substantially as described in the foregoing Example.

10. An expandable thermoplastic composition whenever prepared by the method claimed in any of Claims 7 to 9.

11. A method of preparing a uniformly smooth-surfaced, water vapor-resistant, solvent-resistant structure, which comprises heating the composition claimed in any one of Claims 1 to 6, or 10 in a molding operation to a temperature greater than the softening temperature of said particles and the boiling point of said blowing agent contained therein to cause said composition to expand, fuse and form the desired structure.

12. A foam structure whenever prepared by the method claimed in Claim 11.

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